

and graphitized carbon fibers; alloys and oxides comprising Si, Sn, Al, B, Ge, P, Pb and the like; and nitrides such as  $\text{Li}_3\text{N}$ ,  $\text{Ni}_{3-x}\text{Co}_x\text{N}$  are used.

The liquid organic electrolyte forming the gel electrolyte is prepared by dissolving a solute in an organic solvent. As the solute,  $\text{LiPF}_6$ ,  $\text{LiBF}_4$ ,  $\text{LiClO}_4$ ,  $\text{LiN}(\text{CF}_3\text{SO}_2)_2$ ,  $\text{LiN}(\text{C}_2\text{F}_5\text{SO}_2)_2$  and the like are used. As the organic solvent, cyclic or linear carbonates such as ethylene carbonate, propylene carbonate, butylene carbonate, dimethyl carbonate, diethylcarbonate, ethyl methyl carbonate, methyl propyl carbonate and ethyl propyl carbonate, and cyclic or linear esters such as  $\gamma$ -butyrolactone and ethyl propionate are used. The solutes and the organic solvents may be used solely or in combination of two or more.

In the polymer battery, by charging and discharging, lithium ions dissolved in the solvent move between the positive electrode and the negative electrode. If a material which easily swells in the liquid organic electrolyte is used as the binder, the organic solvent moves into the electrodes together with the move of lithium ions, thereby to swell the electrode plates. In particular, in the negative electrode, the electrode plate greatly swells due to the organic solvent moving at the time of charging, which is liable to cause breakage of the electronically conductive network in the electrode plate and a liberation of the active material particles. Consequently, as the binder contained in the

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positive electrode or the negative electrode, desired is one not liable to swell in the organic solvent, having a good ability to bind the active materials and excelling in the bonding property with metal current collectors and in the affinity with the separator comprising the gel electrolyte.

Conventionally, polyvinylidene fluoride or the like has been used in many cases as the binder. In the structure of polyvinylidene fluoride, since fluorine and hydrogen are arranged alternately and each monomer unit is polarized, it is known that dipole-dipole interaction takes place inside the molecule and between the molecules. However, since polyvinylidene fluoride contains fluorine, polyvinylidene fluoride has a small surface energy and cannot be expected so much for chemical binding effect.

In a preferred embodiment of the present invention, for example, a modified polyvinylidene fluoride having an oxygen-containing group and having a molecular weight of 100,000 to 1,000,000, and a modified vinylidene fluoride-hexafluoropropylene copolymer having an oxygen-containing group and having a molecular weight of 100,000 to 1,000,000 are used. These polymers are provided with a chemical binding function by introducing an oxygen-containing group such as a hydroxyl group, carbonyl group and carboxylic group into polyvinylidene fluoride and a copolymer of vinylidene fluoride-hexafluoropropylene. When these binders are used, the bonding force between the metal current corrector and the

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electrode mixture and between the positive electrode and the separator, and the binding force between the particles in the electrode mixture can be increased by the effect of the oxygen-containing group. The oxygen-containing group works effectively when the ratio of the introduced oxygen-containing group is about 0.1 to 2 wt% to the weight of the polymer before modified.

The oxygen-containing group can be introduced into polyvinylidene fluoride or a copolymer of vinylidene fluoride-hexafluoropropylene by the oxidation reaction accompanying dehydrofluoric acid reaction in NMP, as described above. This oxidation reaction is accelerated by the presence of alkaline substances such as lithium hydroxide and amines. A modified polyvinylidene fluoride having an oxygen-containing group and a modified vinylidene fluoride-hexafluoropropylene copolymer having an oxygen-containing group are available from several manufactures. As commercially available materials, MKB polymer manufactured by ATOFINA CHEMICALS, Inc. (formerly, Elf atochem), #9130 manufactured by Kureha Chemical Industry Co., Ltd. and the like can be mentioned.

The lithium-containing complex oxide in the positive electrode is readily mixed with alkaline impurities. Therefore, from the viewpoint of suppressing the dehydrofluoric acid reaction accompanying the viscosity increase of the positive electrode active material, it is preferable to use a modified vinylidene fluoride-

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